



IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicants : Miyoshi et al.
Serial No. : 10/553,209
Filed : October 13, 2005
For : SHAPED RESIN ARTICLE AND CONDUCTIVE RESIN
COMPOSITION
Art Unit : 1796
Examiner : Ana Lucrecia Woodward

DECLARATION UNDER 37 C.F.R. 1.132

I, Takaaki Miyoshi, a Japanese citizen residing at 1711-2 Hitomi, Kimitsu-shi, Chiba-ken, Japan, declare and say:

I took a major in Industrial Chemistry at Kobe City College of Technology, and graduated therefrom in March 1987.

In April 1987, I entered Asahi Kasei Kogyo Kabushiki Kaisha (now, Asahi Kasei Kabushiki Kaisha), and have been engaged in the research and development of polymer alloys.

I am one of the inventors of the present application and I am well familiar with the present case.

I read and understood the Office Action dated January 6, 2009 and references cited therein.

I have made observations on the necessity of a partially hydrogenated aromatic vinyl/conjugated diene block copolymer having a number average molecular weight of from 200,000 to 300,000 for producing the shaped resin article of

the present invention, with reference to Example 5 and Comparative Example 2 of the present application. The observations are described in a paper attached hereto and marked "Exhibit 1".

From the observations of Exhibit 1, it can be fairly concluded:

(1) that the shaped resin article of the present invention obtained in Example 5 --which comprises a polyamide (A) comprising at least two different polyamide components, a polyphenylene ether (B), and a partially hydrogenated block copolymer (C-1) having a number average molecular weight (Mn) of from 200,000 to 300,000, and which has a polyamide area ratio of at least 80 %-- has an excellent matte surface, whereby the shaped resin article exhibits excellent coating adhesion strength without sacrificing the sharpness of an image reflected in the coating (Observations of item 3 of Exhibit 1);

(2) that, on the other hand, when the above-mentioned block copolymer (C-1) is replaced by a block copolymer having an Mn of less than 200,000 (Comparative Example 2), the matteness of the surface of the shaped resin article becomes poor, thereby leading to poor coating adhesion strength (Observations of item 3 of Exhibit 1);

(3) that, in each of the working examples of Miyoshi et al., a shaped resin article is produced using, as an impact modifier, only a block copolymer having an Mn of less than 200,000 as in the case of Comparative Example 2 of the present application and, hence, the shaped resin articles ob-

tained in the working examples of Miyoshi et al. also have poor matte surfaces and poor coating adhesion strength (Observations of item 4 of Exhibit 1);

(4) that, in addition, Miyoshi et al. have no teaching or suggestion about the importance of the use of a block copolymer having an Mn of from 200,000 to 300,000 and the importance of the polyamide area ratio of at least 80 % (Observations of item 4 of Exhibit 1); and

(5) that, from items (1) to (4) above, it is apparent that the shaped resin article of the present invention can-not be obtained by the technique disclosed by Miyoshi et al.

The undersigned petitioner declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: June 22, 2009


Takaaki Miyoshi

Observations on the necessity of a partially hydrogenated aromatic vinyl/conjugated diene block copolymer having a number average molecular weight of from 200,000 to 300,000 for producing the shaped resin article of the present invention, with reference to Example 5 and Comparative Example 2 of the present application

1. OBJECT OF THE OBSERVATIONS

In the Office Action dated January 6, 2009, the Examiner rejects claims 1-15 of the present application as being obvious over U.S. 7,220,795 (Miyoshi et al.) further in view of U.S. 5,965,655 (Mordecai et al.) Specifically, the Examiner states as follows:

"given that compositions of Miyoshi et al have a multi-phased morphology wherein the polyamide component forms the continuous phase, one having ordinary skill in the art would have expected patentee's shaped articles formed from the composition meeting the material requirements of the present claims, said composition clearly being within the purview of the general disclosure of the reference, to have a surface area feature."

However, contrary to the Examiner's understanding, a shaped resin article of the present application cannot be obtained by the technique disclosed by Miyoshi et al. as explained below.

2. ESSENTIAL FEATURES OF THE PRESENT INVENTION

The shaped resin article of the present invention is advantageous in that the shaped resin article has excellent

coating adhesion strength and a coating formed on the shaped resin article has excellent sharpness of an image reflected therein (see, for example, page 1, line 23 to page 2, line 8 of the present specification).

As can be seen from claim 1 of the present application, the shaped resin article of the present invention comprises one or more partially hydrogenated block copolymers (C) including at least one partially hydrogenated block copolymer (C-1) having a number average molecular weight (Mn) of from 200,000 to 300,000, and the surface area of the polyamide (A) (polyamide area ratio) exposed on the overall surface of said shaped resin article is at least 80 %. The use of such a block copolymer (C-1) having a specific Mn, in combination with the specific polyamide area ratio, contributes to the above-mentioned excellent effects of the present invention. In this connection, it should be noted that the Mn of the block copolymer (C-1) used in the present invention is high as compared to the Mn values of those conventionally used in the art, and the above-mentioned effects achieved by the use of such a high molecular block copolymer (C-1) are completely unexpected and surprising as explained below.

Conventionally, it has been believed that the surface of a shaped resin article (which is to be coated) needs to be smooth in order to improve the sharpness of an image reflected in the coating formed thereon. For example, U.S. 5,965,655 (Mordecai et al.) cited by the Examiner is directed to a resin composition for providing a shaped resin article having a Class A surface, namely a glossy, smooth and polished surface

having a high distinctness of image (Abstract and col.1, lines 23-27 and 30-34 of Mordecai et al.). Therefore, block copolymers having a relatively low molecular weight have been used as impact modifiers because the surface of a shaped resin article becomes uneven when such a high molecular weight block copolymer as used in the present invention is used. However, the smooth surface of a shaped resin article reduces the area of the contact between the surface and the coating formed thereon, thereby lowering the coating adhesion strength.

In this situation, the present inventors have unexpectedly found that, contrary to such conventional knowledge as explained above, when a high molecular weight block copolymer (C-1) is used to form minute concavo-convex portions on the surface of the shaped resin article such that the surface becomes matte, and at least 80 % of the concavo-convex surface is occupied by the polyamide (A), the coating adhesion strength can be improved without sacrificing the sharpness of an image reflected in the coating.

Further, the matte surface of the shaped resin article of the present invention is also advantageous in that it becomes easy to uniformly apply a coating composition on the surface of the shaped resin article so as to form a coating having a uniform thickness (page 86, lines 1 to 7 of the present specification). Furthermore, the matte surface is also advantageous in a case where a shaped resin article is desired to have a matte finish, for example, in order to prevent the shaped resin article from being smudged with fingerprints.

3. OBSERVATIONS BASED ON EXAMPLE 5 AND COMPARATIVE EXAMPLE 2
OF THE PRESENT APPLICATION

According to claim 1 of the present application, the shaped resin article of the present invention comprises the following components:

a polyamide (A) comprising at least two different polyamide components,

a polyphenylene ether (B), and

one or more partially hydrogenated block copolymers (C), each independently obtained by partially hydrogenating an unhydrogenated block copolymer comprising at least one aromatic vinyl polymer block comprised mainly of aromatic vinyl monomer units, and at least one conjugated diene polymer block comprised mainly of conjugated diene monomer units, the partially hydrogenated block copolymers (C) including at least one partially hydrogenated block copolymer (C-1) having a number average molecular weight of from 200,000 to 300,000.

Shaped resin articles comprising the above-mentioned components (A), (B) and (C) were prepared in the working examples of the present specification. For example, in **Example 5**, pellets of a resin composition were produced in substantially the same manner as in Example 1 (page 78, line 10 to page 81, line 15 of the present specification) by using the following raw materials:

PA66-b Polyamide 6,6 having a viscosity number of 130 ml/g, $[\text{NH}_2] = 4.2 \times 10^{-5} \text{ mol/g}$, $[\text{COOH}] = 9.1 \times 10^{-5} \text{ mol/g}$;

PA-MB Polyamide/carbon masterbatch produced in Production Example 3 of the present specification;

PPE-1 Polyphenylene ether powder having a reduced viscosity of 0.52 dl/g; and

SEBS1 Polystyrene/polyethylene butylene/polystyrene block

copolymer, styrene content: 33 %, Mn: 246,000.

Specifically, PA66-b and PA-MB were used as the polyamide (A); PPE-1 was used as the polyphenylene ether (B); and SEBS1 having an Mn in the range of from 200,000 to 300,000 (namely 246,000) was used as the partially hydrogenated block copolymer (C-1) (page 80, lines 1-4; page 91, Table 1 and page 94, Table 2 of the present specification).

On the other hand, in **Comparative Example 2** of the present specification, pellets of a resin composition were produced in substantially the same manner as in Example 5, except that **SEBS2**, which is an SEBS block copolymer having a styrene content of 29 % and an Mn of only 98,500, was used instead of SEBS1 (page 80, lines 4-8; page 91, Table 1 and page 94, Table 2 of the present specification).

The pellets obtained in Example 5 and Comparative Example 2 were evaluated in substantially the same manner as in Example 1 and the results of the evaluation were shown in Table 2 of the present specification. For easy reference, relevant data of Table 2 is excerpted and shown in Table 2' below.

Table 2'

Upstream inlet		Ex. 5	Comp. Ex. 2
Feeder 1	PPE-1 (parts by weight)	38	38
Feeder 2	MAH (parts by weight)	0.3	0.3
Feeder 3	SEBS1 (parts by weight)	12	
	SEBS2 (parts by weight)		12
1st downstream inlet			
Feeder 4	PA66-b (parts by weight)	30	30
	PA-MB (parts by weight)	20	20
Polyamide area ratio		81	75
Coating adhesion strength (number of square coating sections left on the surface of a shaped resin article out of 100 square coating sections)		83	45
Sharpness of an image reflected in the coated surface		B	A
Matteness of the coated surface		III	II

As apparent from Table 2' above, the shaped resin article of Example 5 which comprises the polyamide (A), the polyphenylene ether (B) and the block copolymer (C-1) has a polyamide area ratio of at least 80 % (namely 81 %). The matteness of the coated surface thereof was excellent (i.e., "level III" which means that almost entire surface of the shaped resin article, except for a portion thereof corresponding to a gate of a mold, was matte as described at page 85, lines 21 to 23 of the present specification) and the coating adhesion strength was as high as 83. In addition, the sharpness of an image reflected in the coated surface was satisfactory ("Class B" which means that the outline of the image of the fluorescent light reflected in the coated surface is indistinct but is recognizable as described at page 88, lines 1 to 3 of the

present specification).

On the other hand, the shaped resin article of Comparative Example 2 which comprises the polyamide (A) and the polyphenylene ether (B), but comprises a partially hydrogenated block copolymer having an Mn of less than 200,000 instead of the partially hydrogenated block copolymer (C-1) has a polyamide area ratio of only 75%. Although the sharpness of an image reflected in the coated surface was satisfactory (i.e., "Class A" which means that the outline of the image of the fluorescent light reflected in the coated surface is clearly distinct as described at page 87, lines 23 to 25 of the present specification), the matteness of the coated surface was poor (i.e., "level II" which means that the shaped resin article has a matte portion only at a surface portion thereof corresponding to a point in the mold at which a flow of a molten resin stops as described at page 85, lines 17 to 20 of the present specification) and the coating adhesion strength was also poor (as low as 45).

The reason for the low polyamide area ratio in Comparative Example 2 is not clear but considered as follows.

In Comparative Example 2, due to the low molecular weight of the block copolymer, the melt viscosity of a mixture of the polyphenylene ether (forming the dispersed phase) and the block copolymer is lowered such that the flow of a resin mixture in an injection molding machine causes a large amount of the polyphenylene ether to be exposed on the surface of the resultant shaped resin article.

4. OBSERVATIONS ON MIYOSHI ET AL.

As a component corresponding to the partially hydrogenated block copolymer (C) used in the present invention, Miyoshi et al. describe an aromatic vinyl/conjugated diene block copolymer as an impact modifier, which has an Mn of "10,000 to 500,000, most preferably from 40,000 to 250,000" (col. 8, line 64 to col.9, line 2 of Miyoshi et al.) However, in each of the working examples of Miyoshi et al., only a low molecular weight block copolymer (HTR1, HTR2, HTR3 or HTR4) is used as in Comparative Example 2 of the present application. Specifically, the Mn values of the block copolymers used in the working examples of Miyoshi are: 150,000 (HTR1), 51,000 (HTR2), 51,000 (HTR3) and 120,000 (HTR4) (col. 23, lines 7-49, especially lines 16, 25, 35 and 44 of Miyoshi et al.), all of which are lower than the range (200,000 to 300,000) recited in claim 1 of the present application. This is because, as already mentioned in item 2 above, it has conventionally been believed that a shaped article needs to have a smooth surface to form thereon a coating having excellent sharpness of an image reflected therein.

In the working examples of Miyoshi et al., the materials and process used are not exactly the same as in Comparative Example 2 of the present application. Nevertheless, since only a low molecular weight block copolymer is used in each of the working examples of Miyoshi et al., the resultant shaped resin article has a smooth surface so that a matte surface as desired in the present invention cannot be achieved and the coating adhesion strength of the shaped resin article is poor,

as in the case of Comparative Example 2 of the present application.

Needless to say, Miyoshi et al. have no teaching or suggestion about the importance of the use of a block copolymer having an Mn of from 200,000 to 300,000 and the importance of the polyamide area ratio of at least 80 %.

5. CONCLUSION

From the observations of item 3 above, it is apparent that the shaped resin article of the present invention obtained in Example 5 --which comprises a polyamide (A) comprising at least two different polyamide components, a polyphenylene ether (B), and a partially hydrogenated block copolymer (C-1) having a number average molecular weight (Mn) of from 200,000 to 300,000 and which has a polyamide area ratio of at least 80 %-- has an excellent matte surface, whereby the shaped resin article exhibits excellent coating adhesion strength without sacrificing the sharpness of an image reflected in the coating.

On the other hand, when the above-mentioned block copolymer (C-1) is replaced by a block copolymer having an Mn of less than 200,000 (Comparative Example 2), the matteness of the surface of the shaped resin article becomes poor, thereby leading to poor coating adhesion strength.

As discussed in the observations of item 4 above, in each of the working examples of Miyoshi et al., a shaped resin article is produced using, as an impact modifier, only a block copolymer having an Mn of less than 200,000 as in the case of

Comparative Example 2 of the present application and, hence, the shaped resin articles obtained in the working examples of Miyoshi et al. also have poor matte surfaces and poor coating adhesion strength.

In addition, Miyoshi et al. have no teaching or suggestion about the importance of the use of a block copolymer having an Mn of from 200,000 to 300,000 and the importance of the polyamide area ratio of at least 80 %.

Therefore, it is apparent that the shaped resin article of the present invention cannot be obtained by the technique disclosed by Miyoshi et al.



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United States Patent [19]
Modic

[11] **Patent Number:** **5,723,543**
[45] **Date of Patent:** **Mar. 3, 1998**

[54] **BLOCK COPOLYMERS WITH IMPROVED
OVERMOLDING ADHESION**

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[73] **Assignee:** **Shell Oil Company, Houston, Tex.**

[21] **Appl. No.:** **721,128**

[22] **Filed:** **Sep. 26, 1996**

[51] **Int. Cl.⁶** **C08L 53/02**

[52] **U.S. Cl.** **525/98; 524/504; 524/525;
524/94; 524/95; 524/99**

[58] **Field of Search** **525/98, 94, 95,
525/99**

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[57] **ABSTRACT**

A thermoplastic elastomer composition is provided. The composition comprises a hydrogenated elastomeric block copolymer having at least two resinous endblocks of polymerized monovinyl aromatic compound and an elastomeric midblock of polymerized conjugated diene, a midblock plasticizer, and an ethylene-acrylate copolymer or terpolymer. The thermoplastic elastomer exhibits improved overmolding adhesion to polar matrices over compositions not containing the ethylene-acrylate polymer.

26 Claims, No Drawings

**United States Patent** [19]**Troska et al.**[11] **Patent Number:** **5,932,648**[45] **Date of Patent:** **Aug. 3, 1999**[54] **LOW VOC, HIGH SOLIDS FUMIGATION
ADHESIVE COMPOSITION**[75] **Inventors:** **James Louis Troska**, Costa Mesa,
Calif.; **Larry McArthur Kegley**,
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Dances, Ontario, Calif.[73] **Assignees:** **Shell Oil Company**, Houston, Tex.; **AC**
Products, Placentia, Calif.[21] **Appl. No.:** **08/712,402**[22] **Filed:** **Sep. 11, 1996****Related U.S. Application Data**[60] **Provisional application No.** 60/003,851, Sep. 15, 1995.[51] **Int. Cl.⁶** **C08L 53/02**[52] **U.S. Cl.** **524/505; 524/271; 524/274;**
524/499; 525/98; 525/99[58] **Field of Search** **524/271, 274,**
524/499, 505; 525/98, 99[56] **References Cited****U.S. PATENT DOCUMENTS**

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Primary Examiner—Peter A. Szekely
Attorney, Agent, or Firm—Donald F. Haas[57] **ABSTRACT**

A low viscosity, low VOC content fumigation adhesive formulation which is comprised of

a) 100 parts by weight (pbw) of a blend of a styrene-diene triblock copolymer, hydrogenated or unhydrogenated, having a weight average molecular weight of 30,000 to 200,000 and a polystyrene content of from 10 to 40% by weight of the polymer, and a styrene-diene diblock copolymer having a weight average molecular weight of 10,000 to 40,000 and a polystyrene content of from 10 to 40% by weight of the polymer, wherein the diblock comprises from 60 to 95% by weight of the polymer blend,

b) 75 to 275 parts by weight (pbw) of a tackifying resin which is compatible with the diene blocks, and

c) 50 to 200 parts by weight (pbw) of a solvent for the polymer.

In its preferred embodiment, the invention is the use of SI diblock polymer with a much smaller amount of conventional SEBS or SEPS triblock polymer to produce an extremely low viscosity product which can be processed in the type of equipment presently in use by fumigation adhesive manufacturers and satisfies the VOC standard of 250 grams per liter or less.

34 Claims, No Drawings